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A major aim of this program was to establish computational techniques based on statistical methodologies for the compositional design of structural ceramics. For the purposes of this study we are focusing on silicon nitride based ceramics. The enhancement of properties in silicon nitride based ceramics has come through a combination of controlling chemistry and microstructure. Since no single ab-initio theory can address all relevant properties we had to first establish a workable database from which we could then begin to apply statistical techniques to aid in a design strategy for materials chemistry. The work to date has resulted in

The establishment of a digital library and data warehousing for silicon nitride
Application of multivariate statistics tools on these data sets
Testing of these methods for their predictive capabilities
Developing a microanalytical database on grain boundary chemistry
Identification of the role of additive chemistry on the sintering and fracture toughness characteristics of silicon nitride

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CHEMICAL DESIGN OF STRUCTURAL CERAMICS

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Introduction

A major aim of this program was to establish computational techniques based on statistical methodologies for the compositional design of structural ceramics. For the purposes of this study we are focussing on silicon nitride based ceramics. The enhancement of properties in silicon nitride based ceramics has come through a combination of controlling chemistry and microstructure. Since no single ab-initio theory can address all relevant properties we had to first establish a workable database from which we could then begin to apply statistical techniques to aid in a design strategy for materials chemistry. The work to date has focussed on four major efforts:

- The establishment of a digital library and data warehousing for silicon nitride
- Application of multivariate statistics tools on these data sets
- Testing of these methods for their predictive capabilities
- Developing a microanalytical database on grain boundary chemistry

Summary Of Key Findings

- For instance, the density of engineering ceramics is influenced by innumerable parameters that have been both empirically as well as theoretically shown to have an influence on this property. These range from bonding influences of small amounts of chemical additives, microstructure, sintering conditions and many other combinations of process variables. As shown below, if one maps the vast array of combinations of parameters or descriptors affecting this property, the challenge is immense.

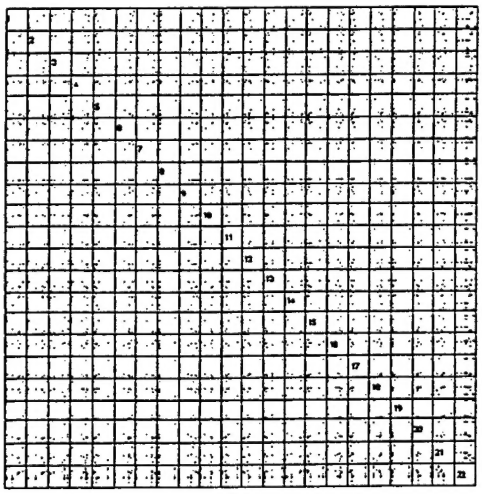

Figure 1: Combinatorial response map of 22 descriptors in silicon nitride

Based on this "Combinatorial Response Map" which summarizes a vast array of structure-process- chemistry relationships associated with properties in silicon nitride, a detailed principal component analysis was carried out. Based on this PCA analysis, we have demonstrated that we have organized a reasonably robust data mining tool that appears to have some success as a predictive formulation as shown in the figure below.

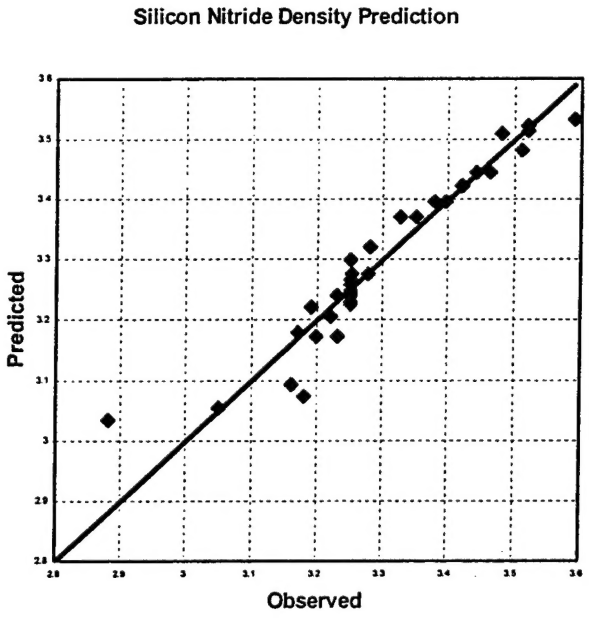

Figure 2: Validation measure of data mining tools

- As shown above, with so many descriptors the question is which ones are in fact the most valuable in discerning their "information content", ie. Which is the most statistically rich in information? The information-theory based concept for entropy was introduced by Shannon as a measure of information in classical digital communication theory. This has been recently applied in the field of bioinformatics to capture descriptor variability independent of value ranges and distributions and to rank the descriptors for selection purposes..

  Shannon's Entropy is defined as

  $$SE = -\Sigma p_i \log_2 p_i$$

  where p is probability of observing a particular descriptor value. p is calculated from the number of compounds with a descriptor value that falls within a specific histogram bin, or "count" (c), for a specific data interval i.

  $$p_i = c_i / \Sigma c_i$$

  Here the logarithm to the base 2 is a scale factor and the SE is a measure of the number of binary bits necessary to capture the information contained within the descriptor variation. The SE values for different descriptors can be directly compared if and only if we use an uniform binning method. Hence the number of data intervals

- In parallel to the establishment of the basic computational tools for having a "Database Engineering" tool, we have also expanded the concept of data to spectral and visual datasets. The aim of this effort is to eventually have a seamless information system whereby one can map microstructure and microchemistry and directly infer possible properties or processing histories through statistical learning tools. A first but critical step in this process is to be able to quantify high resolution chemical information with a pixel by pixel association with the high resolution images. This is being accomplished by developing scatter plots derived from energy loss spectral data where a great deal of care has been taken to ensure that the quantitative aspects of EELS data acquisition have been met. In order to study chemistry-property relationship, we need to know the correlation among those elements, ie. if the concentration change of one element with respect to another. Hence Scatter diagram is introduced to correlate two or more elemental maps to observe the relationship. Once we identify the clusters of concentration distribution, we can also use that information to retrace this pixel-by-pixel information back to the original image so that we can better understand the spatial distribution of the relative chemistries in multicomponent systems.
- The influence of alloying elements on this Silicon Nitride sample is shown below in the scatter diagram as a separation of two clusters, which can be directly related to the EELS spectra: As we move across the grain boundary, the EELS spectra of Nitrogen K-edge shows significant changes. However, when we correlate the variation of the chemical structures of both Silicon and Nitrogen, as in this scatter diagram, it shows two different trends for the two clusters: both the range of intensity distribution and the slope of the major axis of the contour plot. That indicates possible compositional fluctuations for the alloying elements in those two grains, as well as the behavior variation of silicon nitride at the grain boundary.

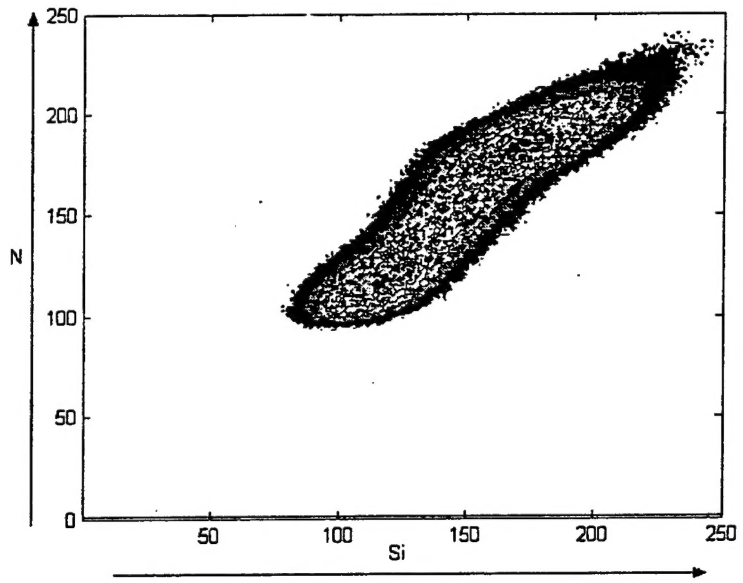

Figure 4: EELS grain boundary chemistry scatter plot

Summary

- Developed digital library on silicon nitride mechanical properties – foundation for the first functional data base for materials design
- Established working multivariate analysis programs for principal component analysis methods
- Demonstrated robustness and validity of descriptor selection through predictive testing methodologies
- Tested new machine learning algorithms (ie. support vector machines and association mining) on silicon nitride data sets
- Initiated electron microscopy studies on interface chemistry in silicon nitride ceramics and relate the information on chemical gradients to database analysis to explore the possibility of "mapping" local properties in a statistical manner to the images.

Awards / Recognition

- CNRS Visiting Professorship – Institute for Organometallic Chemistry, Universite de Rennes Rennes, France

- Co-chair: Engineering Foundation Workshop : Integrating Materials Science into Engineering Structures and Devices –Lake Arrowhead, CA , November 12-14, 2001

Invited Presentations (June 2001- December 2001)

1. *Materials Discovery Through Data Classification*
   European Society for Combinatorial Science –EURO COMBI 2001
   Budapest, Hungary , July 4th 2001

2. *Integrating Length Scales in Combinatorial Chemistry Screening*
   Chemie du solide et inorganique moleculaire section Seminar Series, CNRS,
   Universite de Renne France – July 10th 2001

3. *Materials Design Through Database Engineering*
   Organometalliaque et catalyse section Seminar Series, CNRS, Universite de
   Renne Rennes France– July 11th 2001

4. *Database Engineering for Structural Ceramics*
   AFOSR Program Review Meeting
   Snowbird, Utah – Aug.20th 2001

5. *Data vs Knowledge: What can we learn from databases ?*
   Design Institute for Physical Property Data meeting
   AIChE Annual Meeting, Reno Nevada- DIPPR – Nov 4th 2001

6. *Combinatorial Materials Science and Informatics: seeking patterns for materials discovery*
   Integrating Materials Science into Engineering Structures and Devices
   Eng. Foundation – Lake Arrowhead CA Nov 11th 2001

7. *QSARs for Materials Science*
   Artificial Intelligence and Combinatorial Materials Science Symposium
   Materials Research Society Fall Meeting – Nov 28th 2001 , Boston MA

Publications – 2001

1. *Application Of Principal Component Analysis To Materials Science Data* Journal of Data Science Changwon Suh, Arun Rajagopalan , Xiang Li and Krishna Rajan (in press)

2. *An Informatics Approach To Interface Characterization: Establishing a "Materials by Design" Paradigm* : Krishna Rajan : Science and Technology of Interfaces eds. S.Ankem and C.S.Pande TMS, Warrendale PA (in press)

3. *QSARs for Materials Science* in Artificial Intelligence and Combinatorial Materials Science MRS proceedings , eds. I.Takeuchi et.al ; Krishna Rajan, Changwon Suh, Arun Rajagopalan and Xiang Li (in press)

4. New web site established: http://www.rpi.edu/~rajank/materialsdiscovery/